

Field-cycle-resolved photoionization in solids

P. A. Zhokhov^{1,2} and A. M. Zheltikov^{1,2,3}

¹*Department of Physics and Astronomy, Texas A&M University, 77843 College Station TX, USA*

²*Russian Quantum Center, 143025 Skolkovo, Moscow Region, Russia*

³*Physics Department, International Laser Center,*

M.V. Lomonosov Moscow State University, 119992 Moscow, Russia

(Dated: February 25, 2014)

The Keldysh theory of photoionization in a solid dielectric is generalized to the case of ultrashort driving pulses. We derive a closed-form solution for the nonadiabatic ionization rate in a transparent solid with a periodic dispersion relation, which reveals ultrafast ionization dynamics within the field cycle and recovers the key results of the Keldysh theory in the appropriate limiting regimes.

In his seminal 1964 paper [1], Keldysh presented his celebrated formulas for photoionization, providing a uniform description of multiphoton and tunneling ionization. Over the next five decades, the Keldysh theory of photoionization has been pivotal to research in laser science, providing a commonly accepted framework for a quantitative analysis of ionization in a remarkable diversity of light-matter interaction phenomena, including laser-induced breakdown [2, 3], high-order harmonic [4] and terahertz [5] generation, as well as filamentation of ultrashort light pulses [6, 7]. While the original Keldysh formulas were intended to describe photoionization in a continuous-wave field, several elegant approaches have been proposed [8–10] in the context of rapidly progressing ultrafast technologies [11] and attosecond science [12], to include the wave-packet nature of ultrashort driver pulses inducing an ultrafast ionization of gases. These approaches help identify new field-cycle-sensitive phenomena in electron tunneling [13, 14] and develop novel experimental methods for all-optical detection of electron tunneling dynamics [15, 16].

Extension of the Keldysh model to the photoionization of solids in the field of ultrashort light pulses encounters additional difficulties. Such an extension not only requires an adequate treatment of broadband driver fields, but also calls for a revision of the model of the electron band structure, as the hyperbolic, Kane-type band model adopted in the Keldysh formalism is not intended for the description of dispersion near the zone edges, which gives rise to serious difficulties in the strong-field regime. Here, we generalize the Keldysh theory of photoionization in solids to the case of ultrashort driving pulses by deriving a closed-form solution for the nonadiabatic ionization rate in a transparent solid, which can be used not only to calculate the probability of ionization in the wake of the pulse and after each field cycle, but also to analyze the behavior of the ionization rate within the field cycle. Our analysis presented below in this paper reveals ultrafast ionization dynamics within the field cycle and recovers the results of the Keldysh theory within its range of applicability.

Our analysis of ionization in solids is based, similar to

the Keldysh theory [1], on a two-band approximation of the electron band structure. The electron wave functions in the conduction and valence bands are written, following Keldysh [1], in the form of field-dressed, Volkov-type [17] wave functions (atomic units are used throughout the paper):

$$\psi_{c,v}(\vec{p}, \vec{r}, t) = u_{c,v}(\vec{p}(t), \vec{r}) e^{i\vec{p} \cdot \vec{r} - i \int_{-\infty}^t \mathcal{E}_{c,v}(\vec{p}(\tau)) d\tau}, \quad (1)$$

$$\vec{p}(t) = \vec{p} + \vec{A}(t), \quad (2)$$

where $u_{c,v}(\vec{p}, \vec{r})$ are the Bloch wave functions of the conduction (c) and valence (v) bands, \vec{r} is the position vector, \vec{p} is the crystal quasi-momentum, $\vec{A}(t) = -\int_{-\infty}^t \vec{E}(t_1) dt_1$ is the vector potential, $\vec{E}(t) = \vec{e}E(t)$ is the linearly polarized electric field with polarization direction \vec{e} , and $\mathcal{E}_{c,v}(\vec{p})$ are the energies of conduction (c) and valence (v) bands. Here, unlike the Keldysh theory, the driving field is not assumed to be monochromatic and can have an arbitrary waveform in the time domain instead. Assuming that the valence band is fully occupied and the conduction band is completely empty before the driving field is switched on, we write the probability amplitude for the electron transition to the conduction band (CB) as

$$L(\vec{p}, t) = \mathcal{N} \int_{-\infty}^t V_{cv}(\vec{p}(t')) E(t') e^{-i \int_{-\infty}^{t'} \mathcal{E}(\vec{p}(\tau)) d\tau} dt', \quad (3)$$

where $\mathcal{E}(\vec{p}) = \mathcal{E}_c(\vec{p}) - \mathcal{E}_v(\vec{p})$, $V_{cv}(\vec{p}) = \int u_{c,\vec{p}}^*(\vec{r}) (\vec{r} \cdot \vec{e}) u_{v,\vec{p}}(\vec{r}) d^3\vec{r}$, and \mathcal{N} is the normalization factor.

The population of the conduction band is found as

$$W_c(t) = \int_{BZ} d^3\vec{p} |L(\vec{p}, t)|^2, \quad (4)$$

where the integration is over the first Brillouin zone (BZ).

Up to this point, we have closely followed the derivation by Keldysh [1]. The Keldysh-theory results are recovered from Eqs. (1) – (4) with $\vec{E}(t) = \vec{E}_0 \cos(\omega t)$. The next step, however, will substantially deviate from the Keldysh treatment. In the Keldysh theory, integration

in time in Eq. (3) for a monochromatic laser field is followed by the integration in p in Eq. (4). The approach that we adopt in this work is different, as we integrate over the momentum in Eqs. (3) and (4) first, making no assumption concerning the waveform of the driving field. This change in the order of integration in Eqs. (3) and (4) is central for our analysis, as it helps us to find the population of the conduction band for a driver pulse of general form and to extend the theory to ionization by arbitrarily short light pulses, allowing ionization dynamics within the field cycle to be understood.

To perform integration in \vec{p} in Eq. (4), we need to specify the explicit form of dispersion $\mathcal{E}(\vec{p})$. The Kane-type dispersion used in the Keldysh treatment is known to provide an adequate approximation for the dispersion around the zone center, but fails to describe periodicity of dispersion in the momentum space and dispersion bending near the zone edges. This leads to serious difficulties in the regime of high-intensity fields, when the effects of zone edges may become significant. Here, we address these issues by using a cosine-type dispersion [18, 19],

$$\mathcal{E}(\vec{p}) = \Delta (1 + \alpha - \alpha \cos(d_1 p_1) \cos(d_2 p_2) \cos(d_3 p_3)), \quad (5)$$

where Δ is the band gap, p_1 , p_2 , and p_3 are the projections of the momentum on the Cartesian coordinate axes, and d_1 , d_2 and d_3 are the lattice constants. Parameter α is a measure of the band curvature. It is related to the effective electron-hole mass as $m = (\alpha \Delta (d_1^2 + d_2^2 + d_3^2))^{-1}$.

Introducing $\vec{a} = \{A_1 d_1, A_2 d_2, A_3 d_3\}$ and $\vec{x} = \{p_1 d_1, p_2 d_2, p_3 d_3\}$, where A_1 , A_2 and A_3 are the Cartesian components of the vector potential, we can represent the CB population at time t as

$$W_c(t) = |\mathcal{N}|^2 \int_{-\infty}^t dt_1 \int_{-\infty}^t dt_2 \int_{BZ} d^3 \vec{x} E(t_1) E(t_2) \times V_{cv}(\vec{x} + \vec{a}(t_1)) V_{cv}^*(\vec{x} + \vec{a}(t_2)) e^{-i \int_{t_1}^{t_2} \mathcal{E}(\vec{x} + \vec{a}(\tau)) d\tau}. \quad (6)$$

Since the integrals in t_1, t_2 in Eq. (6) are dominated by the contributions from the saddle points of the oscillating exponent, corresponding to the pole where the matrix element $V(\vec{x} + \vec{a}(t))$ has a residue that is independent of the specific form V as a function of \vec{x} , Eq. (6) can be rewritten as

$$W_c(t) = |\tilde{\mathcal{N}}|^2 \int_{-\infty}^t dt_1 \int_{-\infty}^t dt_2 \int_{BZ} d^3 \vec{x} E(t_1) E(t_2) \times \exp \left\{ -i \int_{t_1}^{t_2} \mathcal{E}(\vec{x} + \vec{a}(\tau)) d\tau \right\}. \quad (7)$$

In a one-dimensional case the integration in dx in Eq.

(7) yields

$$W_c(t) = |\tilde{\mathcal{N}}|^2 \int_{-\infty}^t dt_1 \int_{-\infty}^t dt_2 E(t_1) E(t_2) G_{1d}(t_1, t_2) \quad (8)$$

where

$$G_{1d}(t_1, t_2) = \int_{-\pi}^{\pi} dx e^{-i(1+\alpha)\Delta(t_2-t_1) + i\text{Re}\Phi \cos x - i\text{Im}\Phi \sin x}, \quad (9)$$

$$\Phi = \Delta \alpha \int_{t_1}^{t_2} \exp \{ia(\tau)\} d\tau, \quad (10)$$

and $\tilde{\mathcal{N}}$ is the field-independent normalization factor. Using the integral representation of the Bessel function, $J_0(z) = \frac{1}{2\pi} \int_{-\pi}^{\pi} e^{iz \sin(x)} dx$, we obtain

$$G_{1d}(t_1, t_2) = 2\pi e^{-i\Delta(1+\alpha)(t_2-t_1)} J_0(|\Phi|). \quad (11)$$

Integration in three dimensions gives (see the Supplementary Material for details)

$$W_c(t) = |\tilde{\mathcal{N}}_{3d}|^2 \int_{-\infty}^t dt_1 \int_{-\infty}^t dt_2 E(t_1) E(t_2) G_{3d}(t_1, t_2), \quad (12)$$

$$G_{3d}(t_1, t_2) = e^{-i\Delta(1+\alpha)(t_2-t_1)} \sum_{k=-\infty}^{\infty} \prod_{s=0}^3 J_k(|\Phi_s|) e^{ik \arg \Phi_s} \quad (13)$$

which differs from the 1D result of Eqs. (8),(11) by the numerical factor \mathcal{N}_{3d} and a more complicated form of the $G_{3d}(t_1, t_2)$ function,

$$\Phi_s = \frac{\alpha \Delta}{4} \int_{t_1}^{t_2} e^{-i(a_1 + a_2 + a_3 - 2a_s \sum_{j=1}^3 \delta_{js})} d\tau, \quad (14)$$

where $s = 0, 1, 2, 3$ and δ_{js} are the Kronecker deltas.

Unlike the Keldysh formalism, which integrates over the time in Eq. (3) assuming a continuous-wave field, our approach does not use any assumption on the shape or the pulse width of the laser field, yielding Eqs. (8)–(14), which allow the CB population to be calculated for a laser field of an arbitrary waveform and pulse width. The Keldysh theory calculates the $L(p)$ amplitude, in accordance with Eq. (3), at the first step, followed by integration over the momentum, as prescribed by Eq. (4), thus yielding the field-cycle-averaged ionization rate for a dielectric with a Kane-type dispersion in the presence of a cw laser field. Our approach, on the other hand, integrates over the momentum at the first step for a periodic dispersion relation, which is better suited for the strong-field regime. This procedure yields the two-time $G_{1d(3d)}(t_1, t_2)$ ionization cross-section function, which is used in the second step to calculate, through the integration over the time, the CB population for a laser field of arbitrary waveform and pulse width.

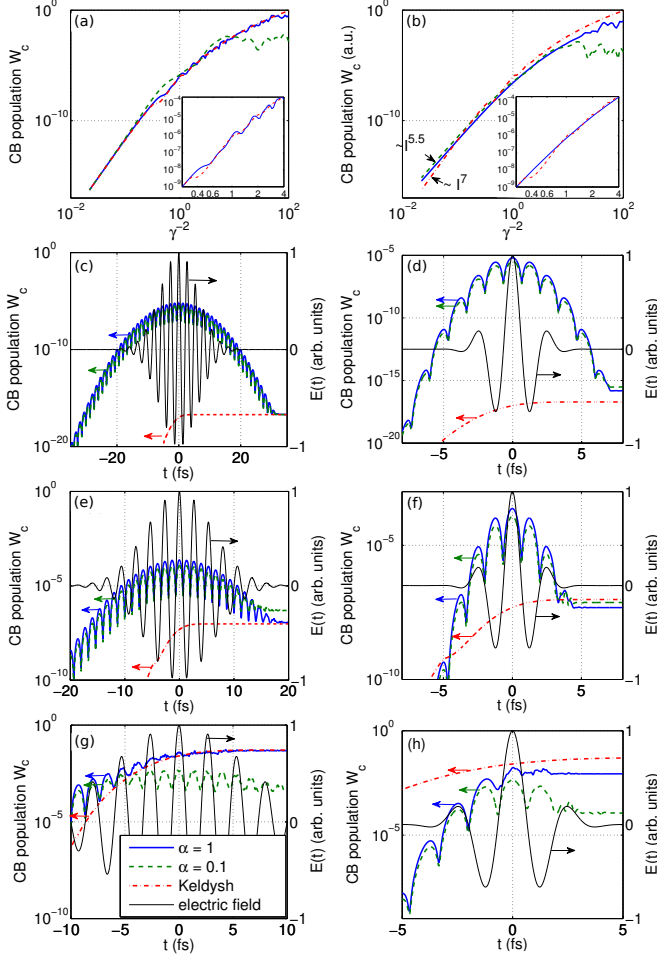


FIG. 1. (a),(b) Population of the conduction band in the wake of the pulse as a function of γ^{-2} with the use of Eqs. (8),(11) with $\alpha = 1$ (solid blue line), with $\alpha = 0.1$ (dashed green line) and using the Keldysh formula (dash-dotted red line). The insets show a close-up of Franz-Keldysh modulation. (c)–(h) Dynamics of the CB population calculated with the use of Eqs. (8), (11) with $\alpha = 1$ (solid blue line) and $\alpha = 0.1$ (dashed green line) and using the Keldysh formula (dash-dotted red line) for $\gamma = 7.2$ (c),(d), $\gamma = 1.2$ (e),(f) and $\gamma = 0.2$ (g),(h). The driver field is shown by the thin black line. The FWHM pulse width is 10 fs (a),(c),(e),(g), 2.4 fs (b),(d),(f),(h), the central wavelength of the driver field is 800 nm, and $\Delta/\omega = 6.45$.

Unlike the periodic dispersion relation of Eq. (5), the Kane-type band model, used in the Keldysh treatment, is not suited to describe the dispersion near the zone edges, failing in the regime of high field intensities where the magnitude of the generalized momentum (2) becomes comparable to reciprocal lattice constants. Predictions of Eqs. (8)–(14) can therefore agree with the Keldysh formula only for relatively low field intensities, where $|x| < \pi/2$, so that the dispersion relation of Eq. (5) can be approximated by a second-order Taylor-series polynomial. In terms of the Keldysh adiabaticity parameter,

$\gamma = \frac{\omega\sqrt{m\Delta}}{E_0}$, where ω is the field frequency, and E_0 is the field amplitude, this condition is written as $\gamma > \frac{2}{\pi\sqrt{\alpha}}$. Furthermore, since the Keldysh formula was derived for a cw field, discrepancies between the predictions of Eqs. (8) – (14) and the Keldysh formula are expected to grow for shorter pulse widths.

Results of calculations presented in Figs. 1(a) – 1(h) fully justify these expectations. For relatively long pulse widths and low field intensities [Figs. 1(a),(c),(e)], when both $\gamma > 1$ and $\gamma > \frac{2}{\pi\sqrt{\alpha}}$ conditions are satisfied, Eqs. (8) – (14) are seen to accurately reproduce the I^k scaling as an asymptotic behavior for the CB population in the wake of the laser pulse as a function of the field intensity I , with k being the minimum number of photons needed to surpass the band gap. However, since the Keldysh theory is not intended for the analysis of ionization on a subcycle time scale, CB population dynamics within each field cycle, as calculations using Eqs. (8) – (14) show, can drastically differ from predictions of the Keldysh formula even in the case of sufficiently long pulse widths and $\gamma > \frac{2}{\pi\sqrt{\alpha}}$ [Figs. 1(c),1(e)]. Specifically, in the regime of low field intensities [Figs. 1(c)–1(f)], the CB populations displays a pronounced oscillatory behavior, following the cycles of the laser field. This oscillatory dynamics within the field half-cycle shows that, in the regime of low field intensities, most of the population transferred from the valence to the conduction band returns back to the valence band within the same field half-cycle, following oscillations of the driver field. When the driver pulse is long enough, however, this oscillatory dynamics converges to the Keldysh theory result in the wake of the laser pulse [Figs. 1(b), 1(c)], indicating the buildup of the multiphoton regime of photoionization as an asymptotic behavior of CB population. Moreover, Eqs. (8) – (14) are seen to accurately reproduce stepwise changes in the CB population as a function of the field intensity [the γ^{-2} parameter in Fig. 1(a)] due to the Franz-Keldysh modulation [20, 21] of the band gap [see the inset in Fig. 1(a)].

It is clearly seen from Fig. 1(a) that the CB population in the wake of the laser pulse calculated with the use of Eqs. (8)–(14) as a function of γ^{-2} (i.e., parameter proportional to the field intensity I) closely follows predictions of the Keldysh theory for $\gamma > \frac{2}{\pi\sqrt{\alpha}}$, but noticeably deviates from the Keldysh theory result when this inequality is not satisfied [e.g., for $\gamma > 2$ in the case of $\alpha = 0.1$ in Fig. 1(a)].

In the case of very short laser pulses, where each field half-cycle is unique, significantly different in its intensity from the adjacent field half-cycles (thin black line in Figs. 1(d), 1(f), 1(h)), the integration over time in Eqs. (8) and (12) no longer converges to the Keldysh theory result even in the wake of the pulse [Figs. 1(b), 1(d), 1(f), 1(h)], showing that the cw-field approximation is no longer applicable even for an asymptotic analysis

of ionization dynamics. Because the number of photons needed for ionization is no longer defined in the regime of very short light pulses, the Franz–Keldysh modulation of the CB population as a function of the field intensity are much less pronounced and are not observed where predicted by the Keldysh formula for a cw field [the inset in Fig. 1(b)].

In the high-intensity regime, $\gamma < 1$, the CB population rapidly builds up after each field half-cycle, giving rise to a stepwise growth of the electron density in the conduction band [Figs. 1(g), 1(h)]. Since the exponential in Eq. (10) is a rapidly oscillating function in this regime, $\Phi(t_1, t_2)$ is vanishingly small unless $|t_2 - t_1| < \epsilon^{-1}$, where $\epsilon = d_1 E$ is the normalized electric field, and d_1 is the lattice constant. We can therefore use a power-series expansion $a(\tau) = a(t_2) - \epsilon(t_2 - \tau)$ to reduce the expression for $G_{1d}(t_1, t_2)$ to

$$G_{1d}(t_1, t_2) = 8\pi \text{Re} \frac{E^2}{\epsilon} e^{-2i\frac{\Delta}{\epsilon}(1+\alpha)\xi} J_0 \left(\frac{2\Delta\alpha}{|\epsilon|} |\sin \xi| \right), \quad (15)$$

where $\xi = \frac{\epsilon}{2}(t_2 - t_1)$.

Replacing the (t_1, t_2) integration variables in Eq. (10) by (ξ, t_2) and using the integral representation of $J_0(x)$, we find for the ionization rate:

$$w(E) = \frac{dW_c}{dt} = 16\pi^2 \frac{E}{d_1} |\mathcal{N}_{1d}|^2 \text{Re} \int_{-\pi}^{\pi} d\eta \int_{-\infty}^{\infty} d\xi e^{-is\nu(\xi, \eta)}, \quad (16)$$

where $s = 2\frac{\Delta\alpha}{\epsilon}$ and $\nu(\xi, \eta) = (\alpha^{-1} + 1)\xi + \sin \xi \sin \eta$.

To simultaneously satisfy the inequalities $\frac{2}{\pi\sqrt{\alpha}} < \gamma < 1$, we require $\alpha > (\frac{\pi}{2})^2$ and calculate the integrals in Eq. (16) using the saddle-point method to derive

$$w(E) = \tilde{K} \frac{E}{s} e^{-s((1+\alpha^{-1})\text{arccosh}(1+\alpha^{-1}) - \sqrt{2\alpha^{-1} + \alpha^{-2}})}, \quad (17)$$

where \tilde{K} is the field-independent numerical constant.

Finally, expanding the exponential in Eq. (17) as a power series in $\alpha^{-1/2}$ up to the first nonvanishing order, we obtain

$$w(E) = K E^2 e^{-\frac{4}{3}\sqrt{\frac{2}{\alpha}}\frac{\Delta}{\epsilon}} = K E^2 e^{-\frac{4}{3}\frac{(2m)^{1/2}\Delta^{3/2}}{E}}, \quad (18)$$

where K is a constant.

It is straightforward to see that Eq. (18) recovers not only the signature exponential, typical of tunneling ionization, but also the E^2 scaling of the pre-exponential factor [20, 22].

We are now in a position to understand effects related to the carrier-envelope phase (CEP) in the photoionization of solids by ultrashort laser pulses. To this end, we represent the driver field as $E(t) = E_0 e^{-(t/T)^2} \cos(\omega t + \phi)$, where T is the pulse duration, and examine the CB population as a function of the CEP ϕ . In the case of long laser pulses, containing many field cycles, i.e., in the regime where Eqs. (8)–(14) recover the

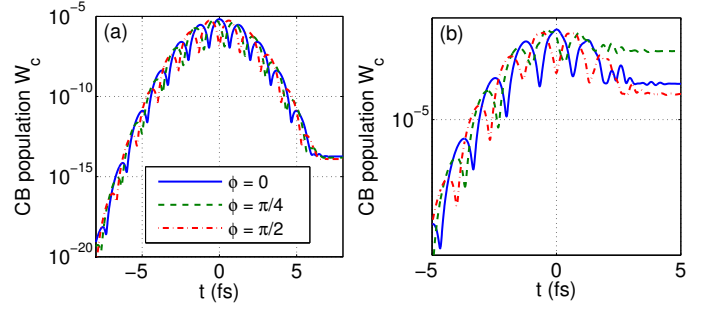


FIG. 2. Dynamics of the CB population for $\gamma = 5$ (a) and 0.2 (b) calculated using Eqs. (8),(11) for the CEP $\phi = 0$ (dashed blue line), $\pi/4$ (solid green line), $\pi/2$ (dash-dotted red line). The FWHM pulse width is 2.4 fs, the central wavelength of the driver field is 800 nm, $\Delta/\omega = 6.45$, $\alpha = 0.1$.

results of the Keldysh theory for cw fields, no CEP dependence is observed, in full agreement with the Keldysh theory.

For very short laser pulses of low intensity, the instantaneous CB population within the field half-cycle is sensitive to the CEP [Fig. 2(a)]. However, the CB population left in the wake of the driver pulse is virtually CEP-independent [$t > 6$ fs in Fig. 2(a)], with almost no deviation from the Keldysh theory. In the regime of high field intensities [Fig. 2(b)], the CB density in the wake of the pulse can be represented as a sum of populations transferred to the conduction band by each field half-cycle [Fig. 2(b)]. The CB population induced by a single field half-cycle, in its turn, is a strongly nonlinear function of the field intensity achieved within this half-cycle. As a result, the CB population in the wake of a very short driver pulse is efficiently controlled by the CEP of this pulse, changing by an order of magnitude in Fig. 2(b) as the CEP is shifted by $\pi/4$.

To summarize, we have extended the Keldysh theory of photoionization of semiconductors to the case of ultrashort driver pulses of arbitrary waveform and pulse width. We derived a closed-form solution for the nonadiabatic ionization rate in a transparent solid, which can be used not only to calculate the probability of ionization in the wake of the pulse, but also to examine ultrafast ionization dynamics within the field cycle. Our approach has been shown to accurately recover the results of the Keldysh theory within its range of applicability.

-
- [1] L. V. Keldysh, Sov. Phys. JETP **20**, 1307 (1965).
 - [2] N. Bloembergen, IEEE Journal of Quantum Electronics **10**, 375 (1974).
 - [3] M. Lenzner, J. Krüger, S. Sartania, Z. Cheng, C. Spielmann, G. Mourou, W. Kautek, and F. Krausz, Physical Review Letters **80**, 4076 (1998).
 - [4] T. Brabec and F. Krausz, Reviews of Modern Physics

- 72**, 545 (2000).
- [5] M. Tonouchi, *Nature Photonics* **1**, 97 (2002).
 - [6] A. Couairon and A. Mysyrowicz, *Physics Reports* **441**, 47 (2007).
 - [7] L. Bergé, S. Skupin, R. Nuter, J. Kasparian, and J.-P. Wolf, *Reports on Progress in Physics* **70**, 1633 (2007).
 - [8] A. M. Perelomov, V. S. Popov, and M. V. Terent'ev, *Sov. Phys. JETP* **23**, 924 (1966).
 - [9] M. V. Ammosov, N. B. Delone, and V. P. Krainov, *Sov Phys JETP* **64**, 4 (1986).
 - [10] G. Yudin and M. Ivanov, *Physical Review A* **64**, 6 (2001).
 - [11] E. Goulielmakis, V. S. Yakovlev, A. L. Cavalieri, M. Uiberacker, V. Pervak, A. Apolonski, R. Kienberger, U. Kleineberg, and F. Krausz, *Science (New York, N.Y.)* **317**, 769 (2007).
 - [12] P. B. Corkum and F. Krausz, *Nature Physics* **3**, 381 (2007).
 - [13] M. Uiberacker, T. Uphues, M. Schultze, A. J. Verhoef, V. Yakovlev, M. F. Kling, J. Rauschenberger, N. M. Kabachnik, H. Schröder, M. Lezius, K. L. Kompa, H.-G. Muller, M. J. J. Vrakking, S. Hendel, U. Kleineberg, U. Heinzmann, M. Drescher, and F. Krausz, *Nature* **446**, 627 (2007).
 - [14] T. Balciunas, A. J. Verhoef, A. V. Mitrofanov, G. Fan, E. E. Serebryannikov, M. Y. Ivanov, A. M. Zheltikov, and A. Baltuska, *Chemical Physics* **414**, 92 (2013).
 - [15] A. J. Verhoef, A. V. Mitrofanov, E. E. Serebryannikov, D. V. Kartashov, A. M. Zheltikov, and A. Baltuska, *Physical Review Letters* **104**, 1 (2010).
 - [16] A. V. Mitrofanov, A. J. Verhoef, E. E. Serebryannikov, J. Lumeau, L. Glebov, A. M. Zheltikov, and A. Baltuska, *Physical Review Letters* **106**, 1 (2011).
 - [17] D. M. Volkov, *Zeitschrift fuer Physik* **94**, 250 (1935).
 - [18] V. Gruzdev, *Physical Review B* **75**, 205106 (2007).
 - [19] S. G. Bonch-Bruевич, V. L. Kalaschnikov, *Halbleiterphysik* (VEB, Berlin, 1982).
 - [20] L. V. Keldysh, *Sov. Phys. JETP* **7**, 763 (1958).
 - [21] W. Franz, *Z. Naturforschung* **13**, 484 (1958).
 - [22] E. O. Kane, *Journal of Physics and Chemistry of Solids* **12**, 181 (1959).